

# Motivation for a range of Ultrafast X-Ray Sources

Roger Falcone

Physics Department, UC Berkeley

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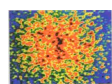
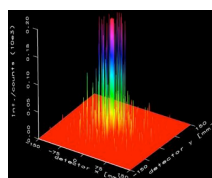
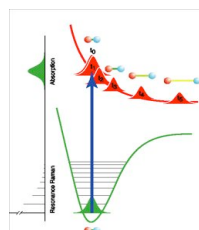
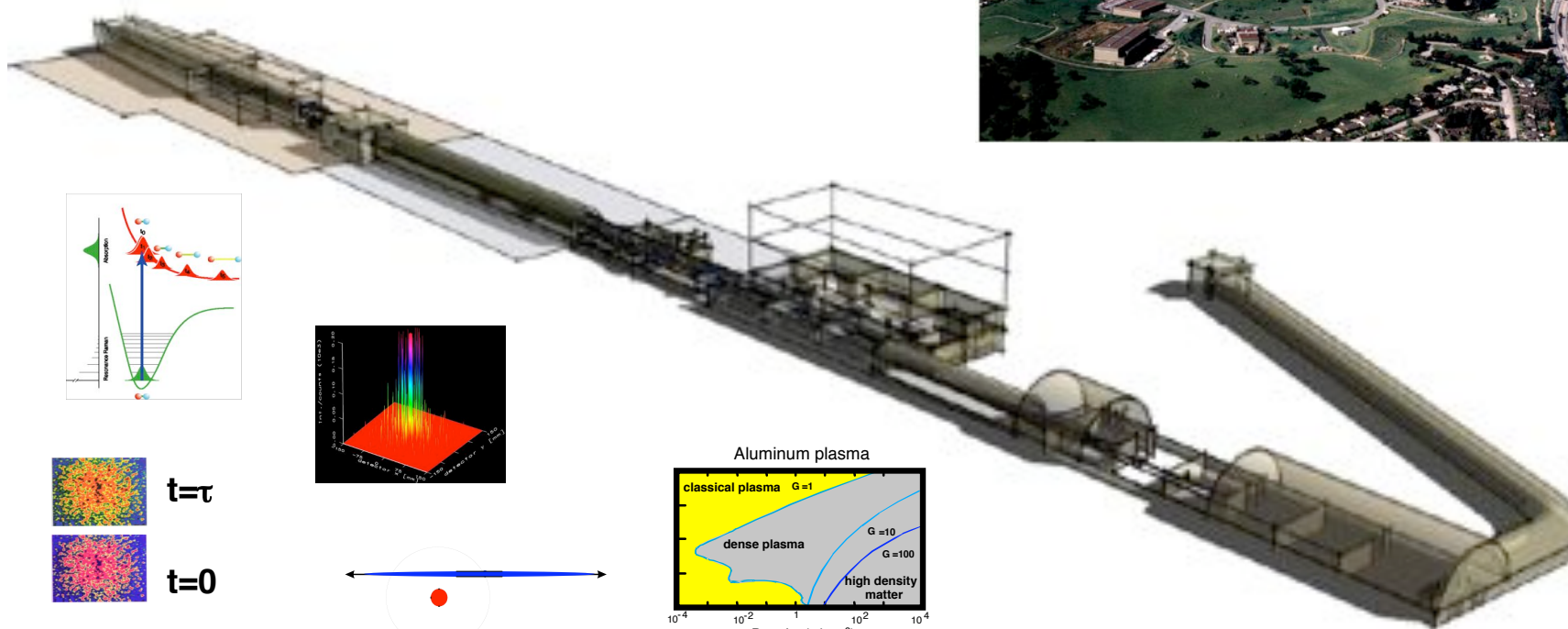
Advanced Light Source  
Lawrence Berkeley National Laboratory

## Observations on ultrafast x-ray science

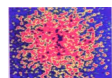
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- workshops and reports, along with successful operation of new sources at APS, ALS, BESSY, SLS, SPPS, FLASH, etc., indicate that compelling ultrafast x-ray science will be enabled by accelerator-based sources
- several new x-ray FELS will soon become available
- optical lasers with high-harmonic-generation and phase-stabilization have indicated the potential of ultrafast, short-wavelength science
- various proposals exist for R&D and construction of new ultrafast x-ray sources, using crabbing, slicing, seeded-FELs, ERLs, lasers, etc.
- high peak and average power lasers will be available for manipulating electron beams and seeding FELs, but accelerator-based x-ray sources have unique capabilities beyond x-ray harmonics of lasers

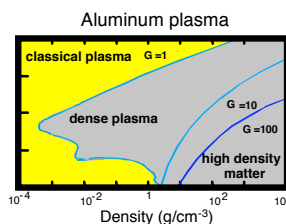
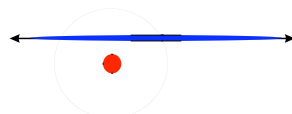
LCLS will enable a range of x-ray science beginning 2009-10



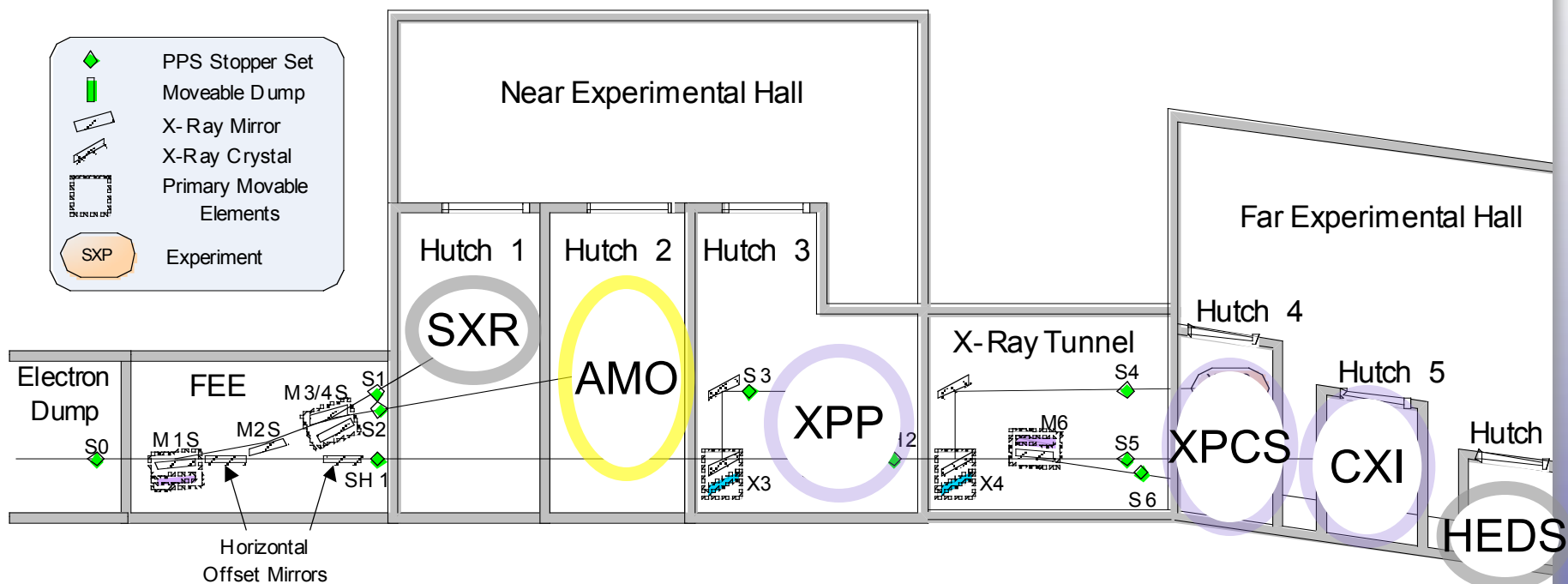
$t=0$



$t=\tau$

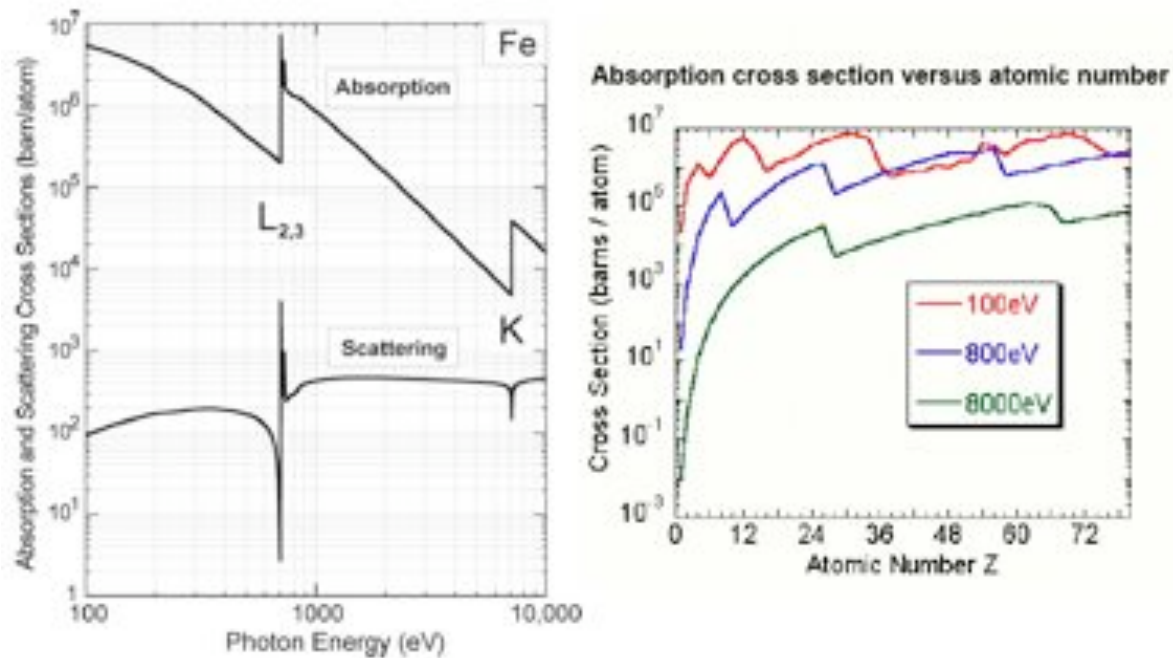


# Near and Far Experimental Hall Hutches for LCLS





## Potential issue: excitation or damage of sample during probing of condensed matter by soft x-ray FELs



$\gamma \sim 10^9 \text{ W @ } 30 \text{ fs @ } 600 \text{ eV} = 3 \times 10^{11} \text{ photons}$

$A \sim (1 - 100 \mu\text{m})^2$

$\sigma \sim 10^{-18} \text{ cm}^2$

$N^* / N = 0.1 - 100\%$

... concerns from Jo Stohr, others



## ...so how many x-rays are too much or too little ?

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- Spontaneous sources

Synchrotron slicing =  $10^2 - 10^4$  photons/pulse at 1-10 kHz

Synchrotron crabbing =  $10^4 - 10^6$  photons/pulse at 6 MHz

SPPS =  $10^8$  photons/pulse at 10 - 100 Hz

- FELs

LCLS =  $10^{12}$  photons at 120 Hz

GW peak power

- Laser harmonics

$10^{-6}$  conversion in peak and average power (MW peak /  $\mu$ W average)

### **A range of new sources and clever techniques are needed**

- amplification of scattering by use of crystals
- dispersive spectroscopy for parallel measurements
- zone plate optics, better detectors, PEEM, diffractive imaging



## Recent Berkeley Workshop "Science for a New Class of Soft X-ray Light Sources"

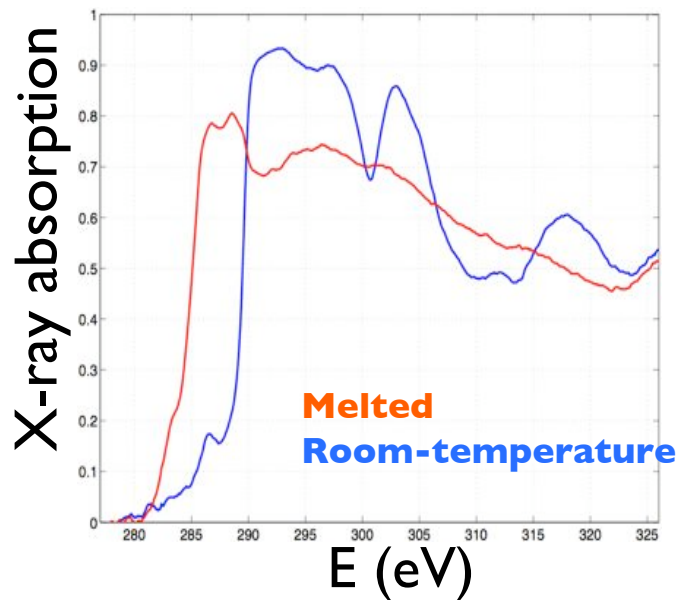
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Focused on 5 areas:

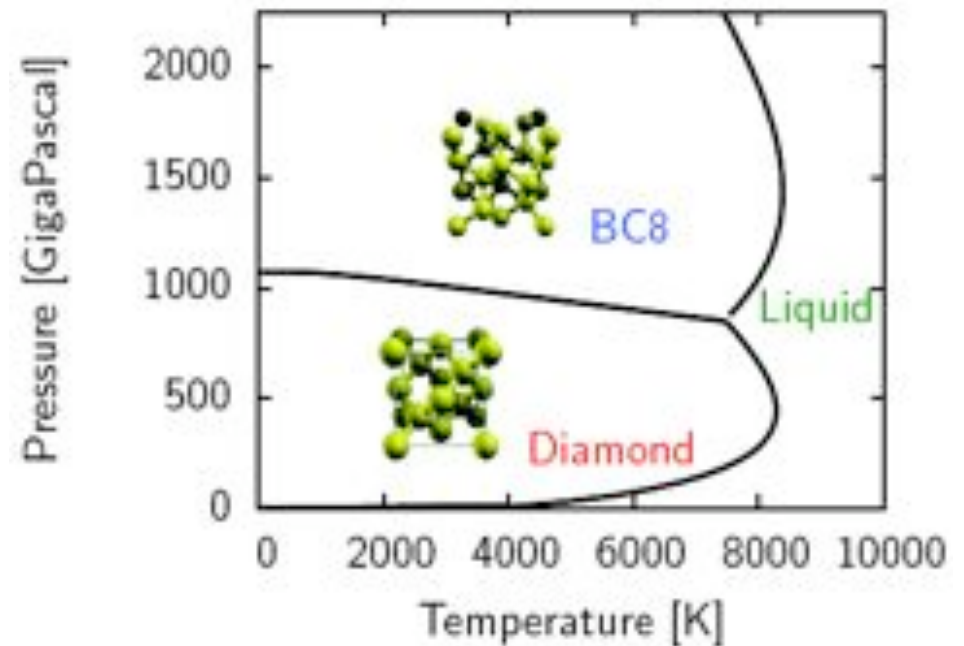
1. Atomic, Molecular and Optical Physics
2. Chemical Physics
3. Correlated Materials
4. Magnetization and Spin Dynamics
5. Nanoscience and Coherence

## Example of dynamic SXR spectroscopy: K-edge absorption of high-T liquid carbon

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Heimann, et al

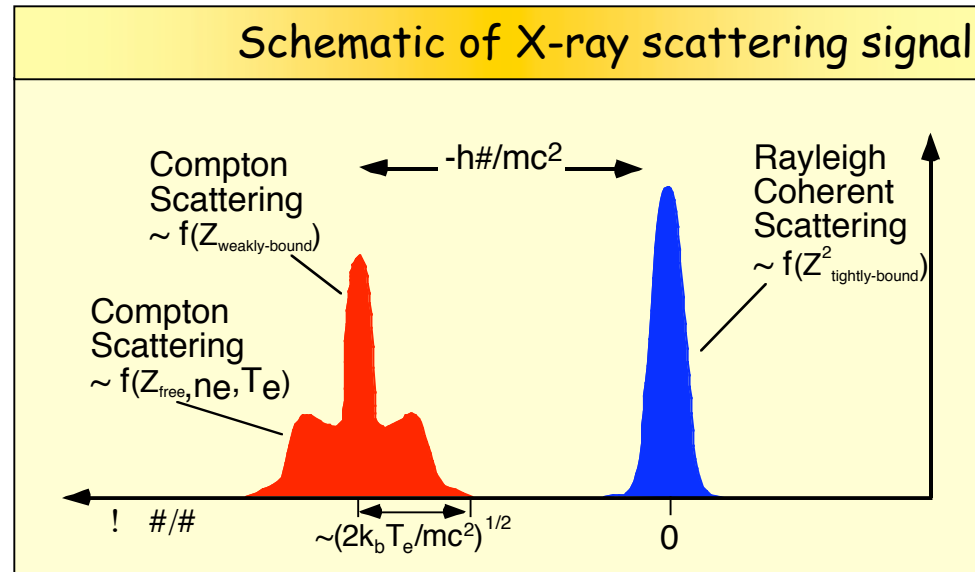


Correa, et al



# Inelastic hard x-ray scattering measures dynamic density and temperature

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**From scattering intensities, we determine:**

- electron motion
- collective motion (plasmons, ion acoustic wave)
- free and bound electrons
- screening and collisions

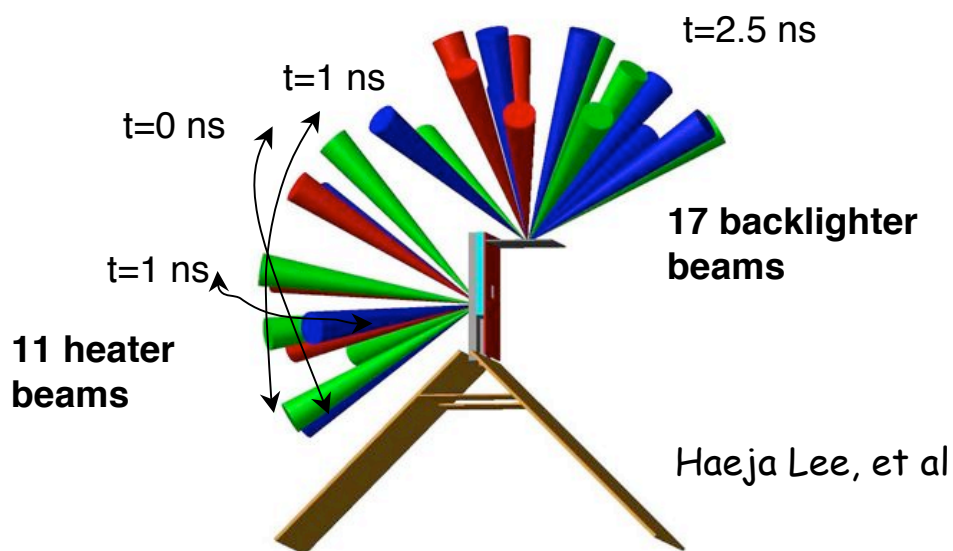
**By varying the scattering angle, we determine:**

- collective modes and non-collective behavior



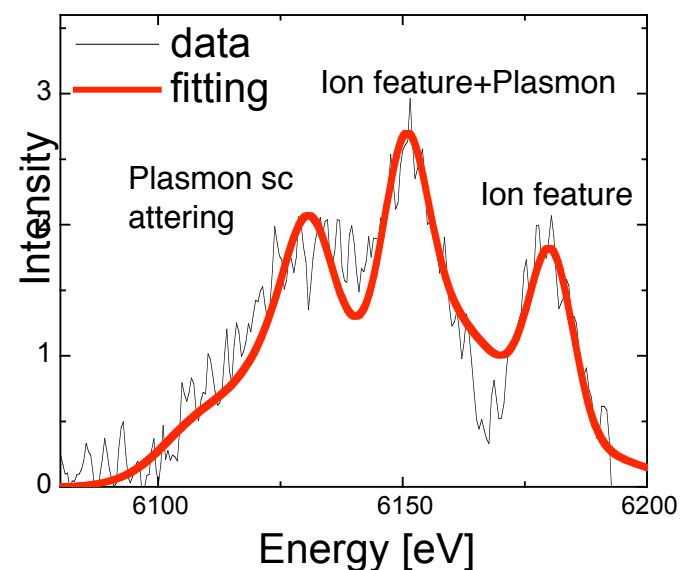
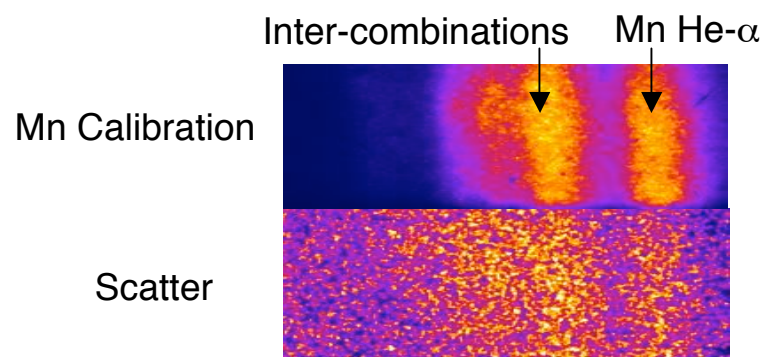
# X-ray Thomson scattering uses x-ray backlighters to study compressed Be

**Laser plasma x-ray source  
used to measure x-ray scattering on compressed Be**

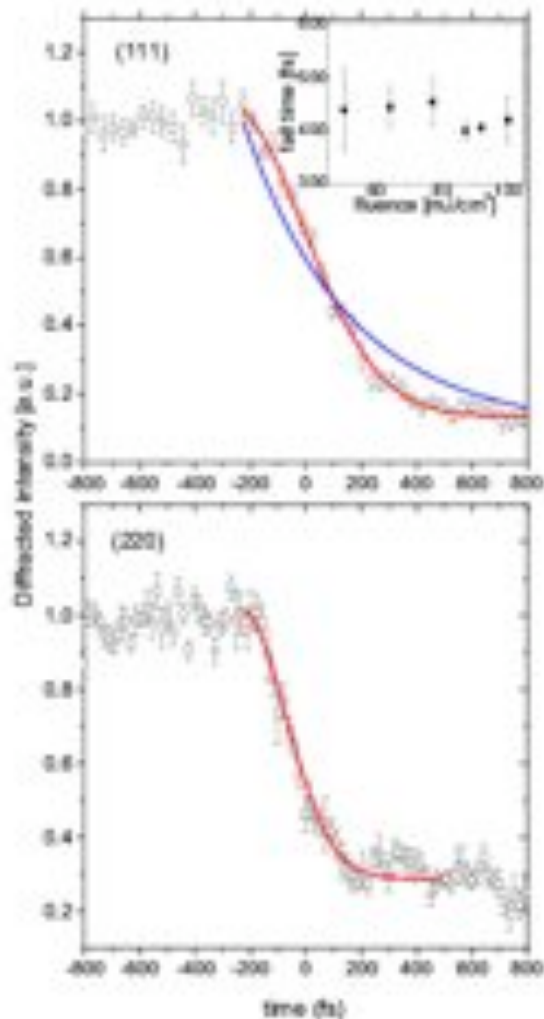


- measured plasmon scattering from shock compressed Be
- position of the plasmon resonance yields density  
 $n_e = 1 \times 10^{23} \text{ cm}^{-3}$ ,  $T_e = 10 \text{ eV}$  at 3 ns

Glenzer, Lee, Falcone, et al



# Disordering of a lattice through bond-breaking observed at short times through 8 keV diffraction changes at the SPPS

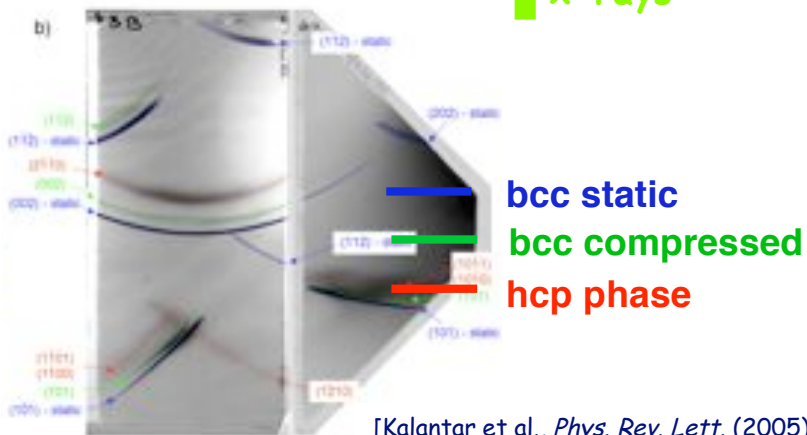
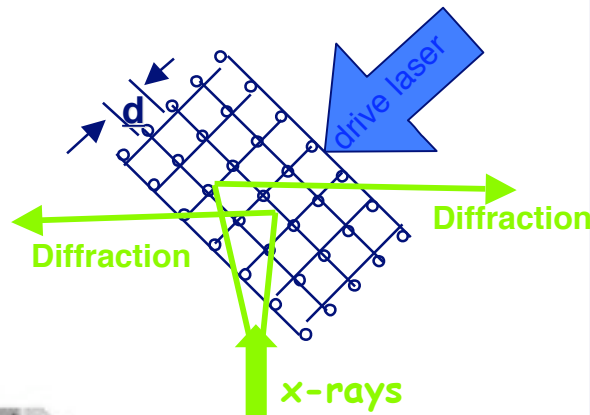


- (111) and (220) reflections measured
- non-thermal melting observed
- more complex system will require more photons

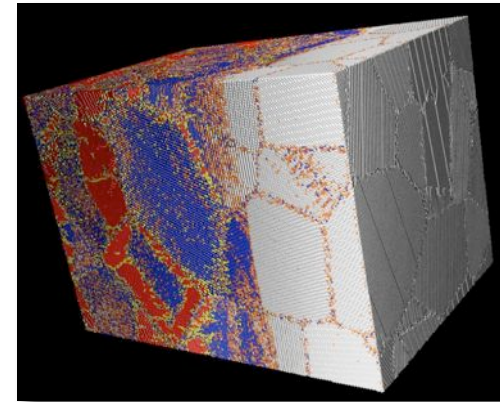
Intense x-ray fluxes from LCLS will enable real-time *in situ* measurements of microstructure evolution at high pressure

## What is the timescale of the bcc-hcp phase transformation in Fe?

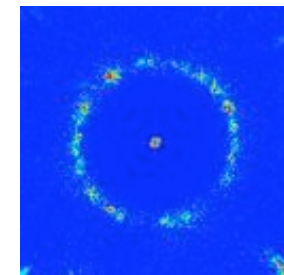
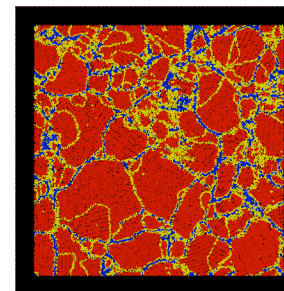
Current measurement  
limited to timescales  $\gg$  psec



Simulations predict subpicosecond phenomena observable using LCLS



Kadau et al., *Science* (2002).





## Recent Berkeley Workshop "Science for a New Class of Soft X-ray Light Sources"

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### Atomic, Molecular and Optical Physics and Chemical Physics Breakouts

- attosecond electron dynamics
- probe and control of electron correlation
- evolution of excited state dynamics in the gas phase
- extreme non-Born-Oppenheimer chemistry
- non-adiabatic control schemes
- 2-d x-ray correlation spectroscopy

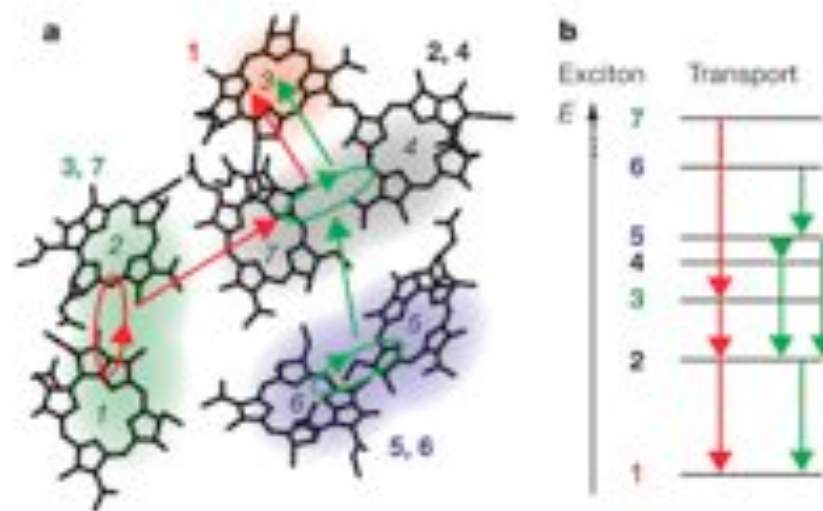
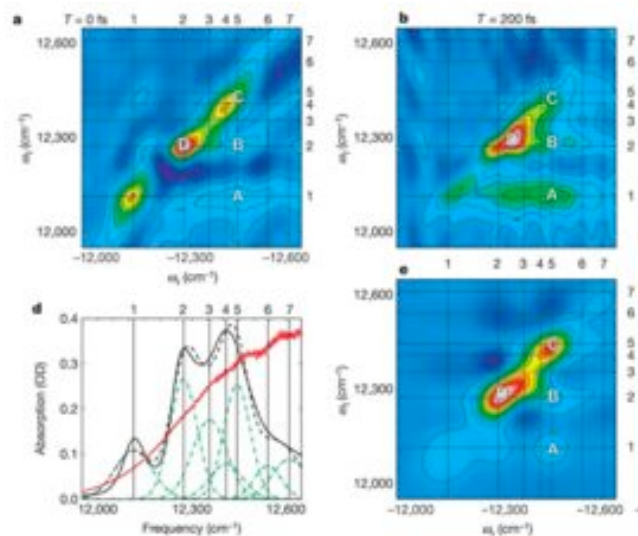
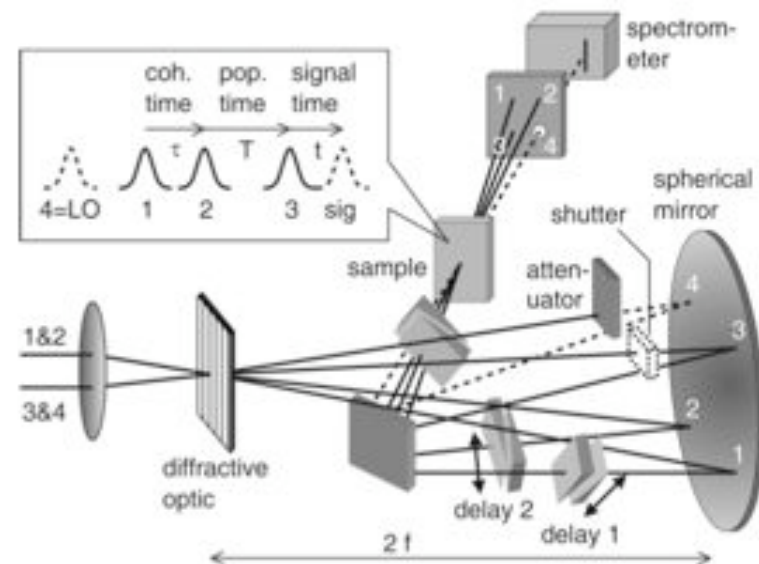
# Two-dimensional spectroscopy of electronic couplings in photosynthesis

Tobias Brixner<sup>1</sup>, Jens Stenger<sup>1</sup>, Harsha M. Vaswani<sup>1</sup>, Minhaeng Cho<sup>2</sup>, Robert E. Blankenship<sup>3</sup> & Graham R. Fleming<sup>1</sup>

<sup>1</sup>Department of Chemistry, and the Institute for Quantitative Biomedical Research (IQBR), University of California, Berkeley, and Physical Biosciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

<sup>2</sup>Department of Chemistry and Center for Multidimensional Spectroscopy, Division of Chemistry and Molecular Engineering, Korea University, Seoul 136-701, Korea

<sup>3</sup>Department of Chemistry and Biochemistry, Arizona State University, Tempe, Arizona 85287, USA





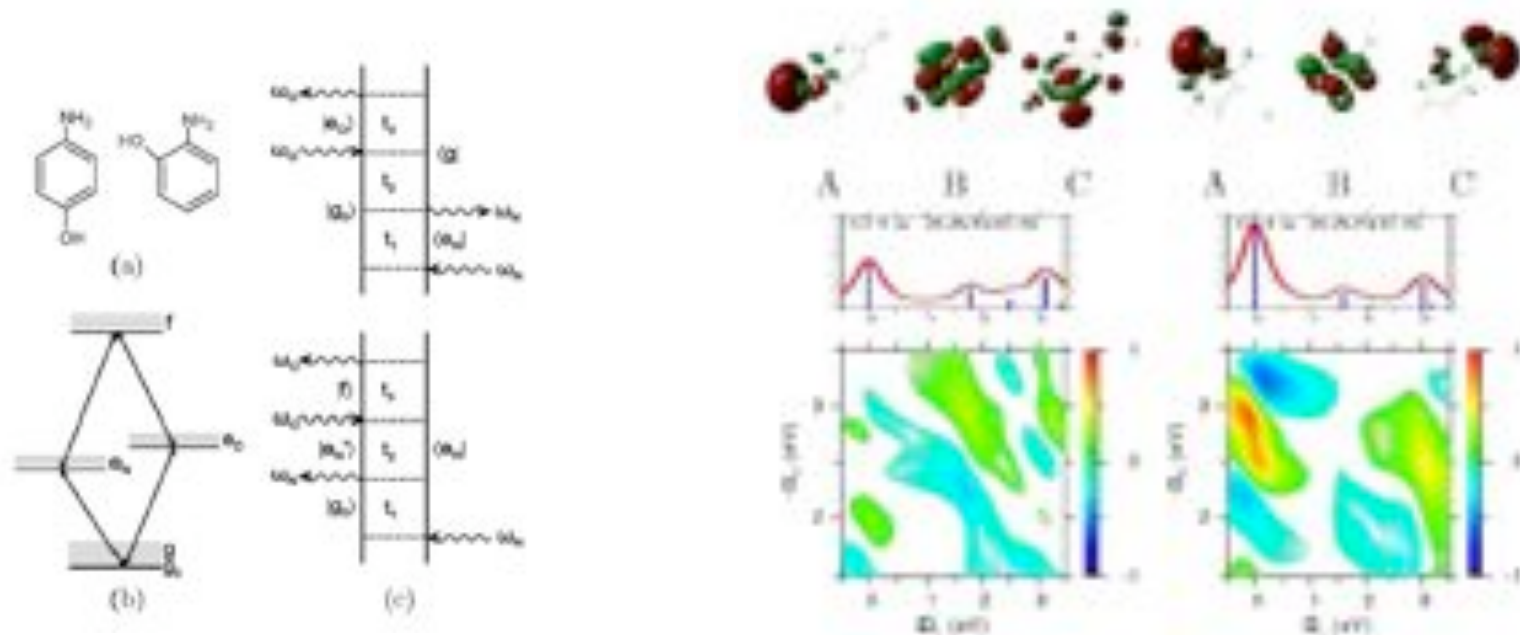
# Coherent ultrafast core-hole correlation spectroscopy; x-ray analogues of multidimensional NMR

Igor V. Schweigert and Shaul Mukamel

Department of Chemistry, University of California, Irvine, California 92697-2025

We propose two dimensional x-ray coherent correlation spectroscopy (2DXCS) for the study of interactions between core-electron and valence transitions. This technique might find experimental applications in the future when very high intensity x-ray sources become available. Spectra obtained by varying two delay periods between pulses show off-diagonal cross-peaks induced by coupling of core transitions of two different types. Calculations of the N1s and O1s signals of aminophenol isomers illustrate how novel information about many-body effects in electronic structure and excitations of molecules can be extracted from these spectra.

PACS numbers: 33.20.Rm, 42.65.Re





## Workshop Conclusions "Science for a New Class of Soft X-ray Light Sources"

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- Function relies on structure, bonding, and dynamics
  - soft x-rays reveal bonding and structure
  - hard x-rays reveal atomic positions
- Energy and information flow utilize ultrafast timescales
  - beat timescales for dissipation  
e.g., vision, photosynthesis
  - allow multimode excitation to dissipate energy  
e.g., DNA, damage
  - speed, competing rates, and quantum pathways  
critical to functional optimization
- Coherent radiation implies longitudinal and transverse coherence for
  - high resolution spatial imaging and spectroscopy
  - high peak and high average power for non-linear measurements
- Imaging matter and energy flow will utilize additional IR to x-ray radiation





# Requirements for new light sources are challenging

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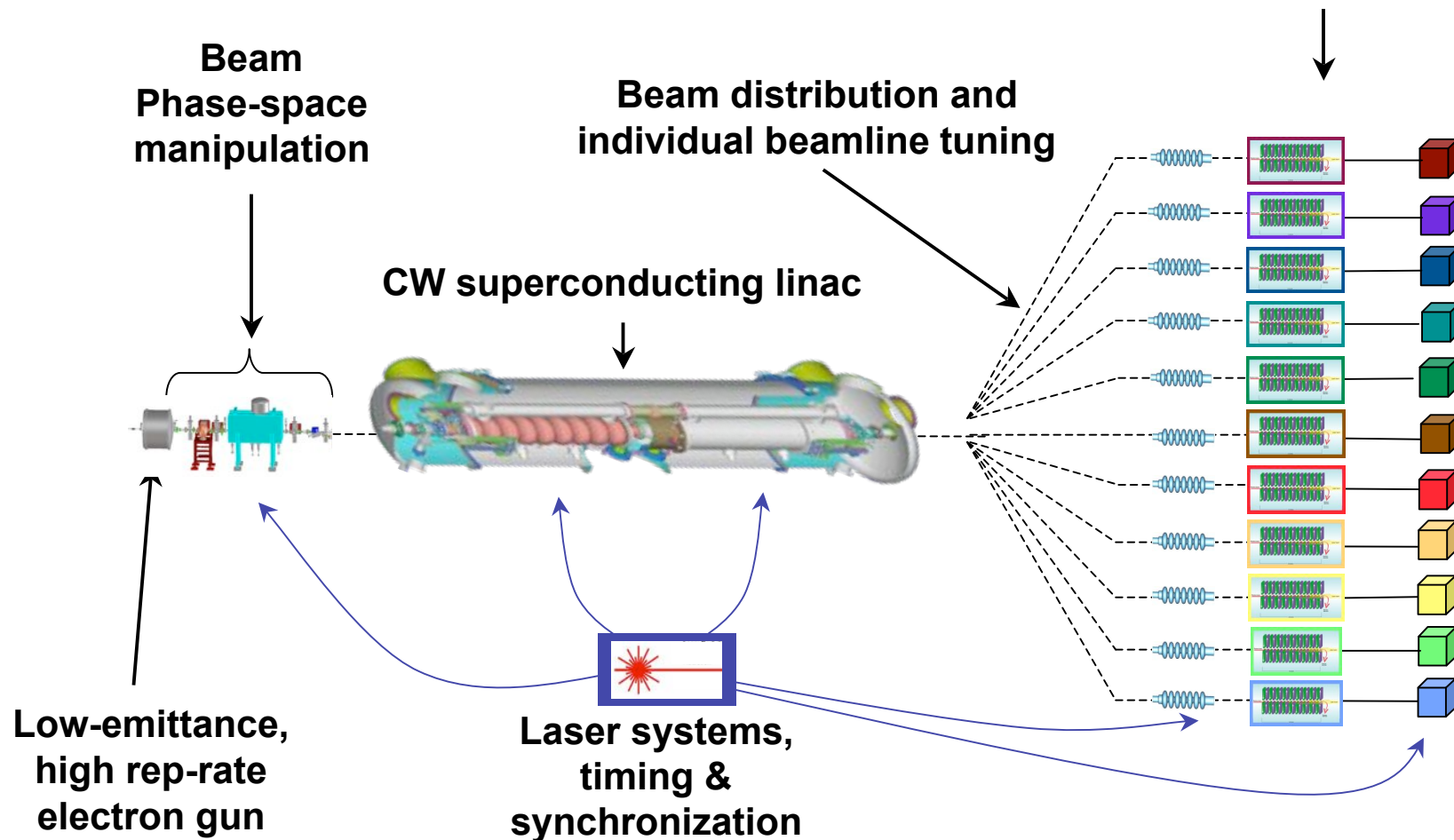
- **Tunable**
  - Spectroscopy, imaging, and near resonance scattering
- **Selectable pulse length**
  - Attosecond science for timescales of electron-electron correlation
  - Longer (time-BW-limited) pulses required for high resolution
- **Selectable pulse energy and repetition rate**
  - Maximize data and S/N, and minimize damage
- **Multiple wavelengths and precision delays**
- **Beam quality**
  - Coherence and stability
- **Amplitude and phase controlled pulses**
- **Synchronized pumps**
  - THz, IR, optical, magnetic - to drive non-equilibrium structures
  - Pressure, temperature, electronic, and phonon excitation beyond ambient
- **Induce nanoscale structure with transient gratings**



# A vision for a future light source facility

## HIGH REP-RATE, SEEDED, VUV — SOFT X-RAY FEL ARRAY

- Independent array of configurable FELs
- Control of electrons: seeded, attosecond, ESASE
- Control of x-rays: wavelength, pulse duration, polarization





# Performance goals of a SXR FEL

## FELs WITH THREE MODES OF OPERATION

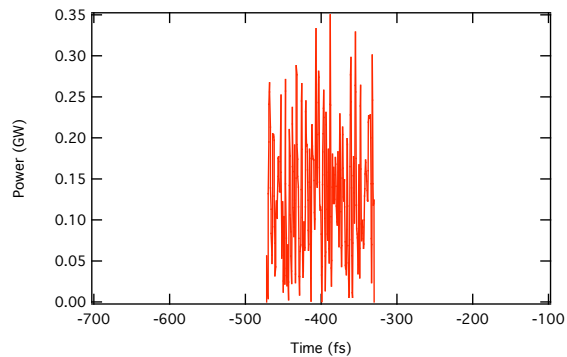
	Short-pulse beamlines	High-resolution beamlines	Sub-femtosecond beamlines
Wavelength range (nm)	~200 – 1	~200 – 1	~40 – 1
Photon energy (eV)	6 – 1240	6 – 1240	30 – 1240
Repetition rate (kHz)	100	100	1-100
Peak power (GW)	1	1	0.1 – 0.3
Photons/pulse (@1 nm)	$5 \times 10^{11}$ (in 100 fs)	$2.5 \times 10^{12}$ (in 500 fs)	$1.5 \times 10^8$ (in 100 as)
Timing stability (fs)	10	10	TBD
Pulse length (fs)	1 – 100	100 – 1000	0.1 - 1
Harmonics	! few%	! few%	! few%
Polarization	Variable, linear/circular	Variable, linear/circular	Variable, linear/circular



# Seeded FEL

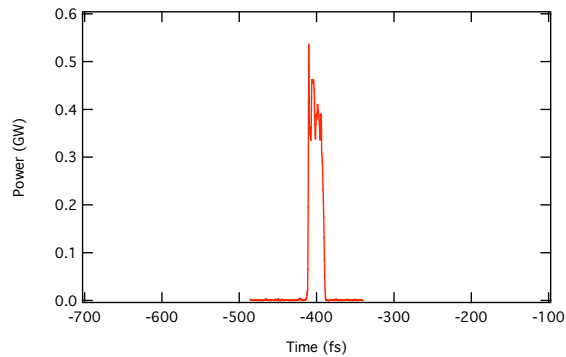
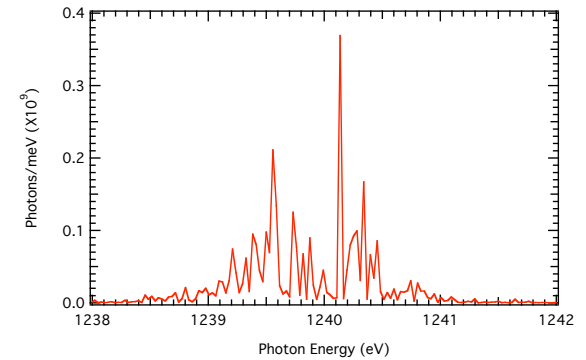
## ENHANCED CAPABILITIES FOR CONTROL OF X-RAY PULSE

### Pulse profile

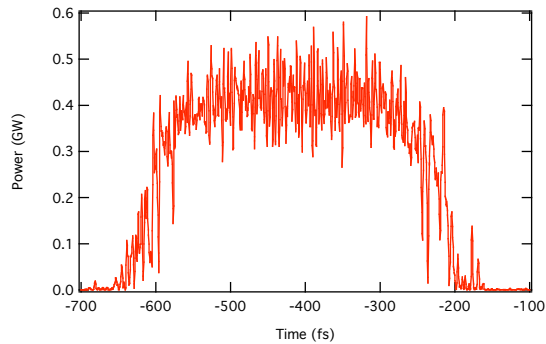
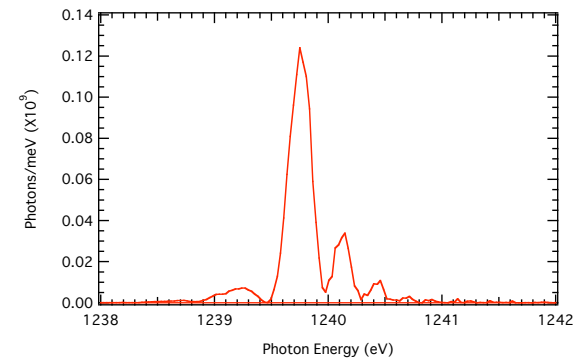


**SASE**

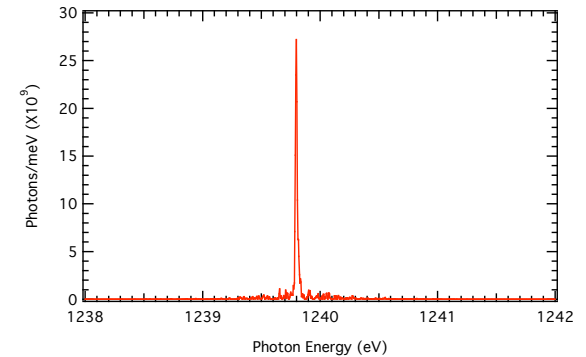
### Spectrum



**25 fs seed**



**500 fs seed**

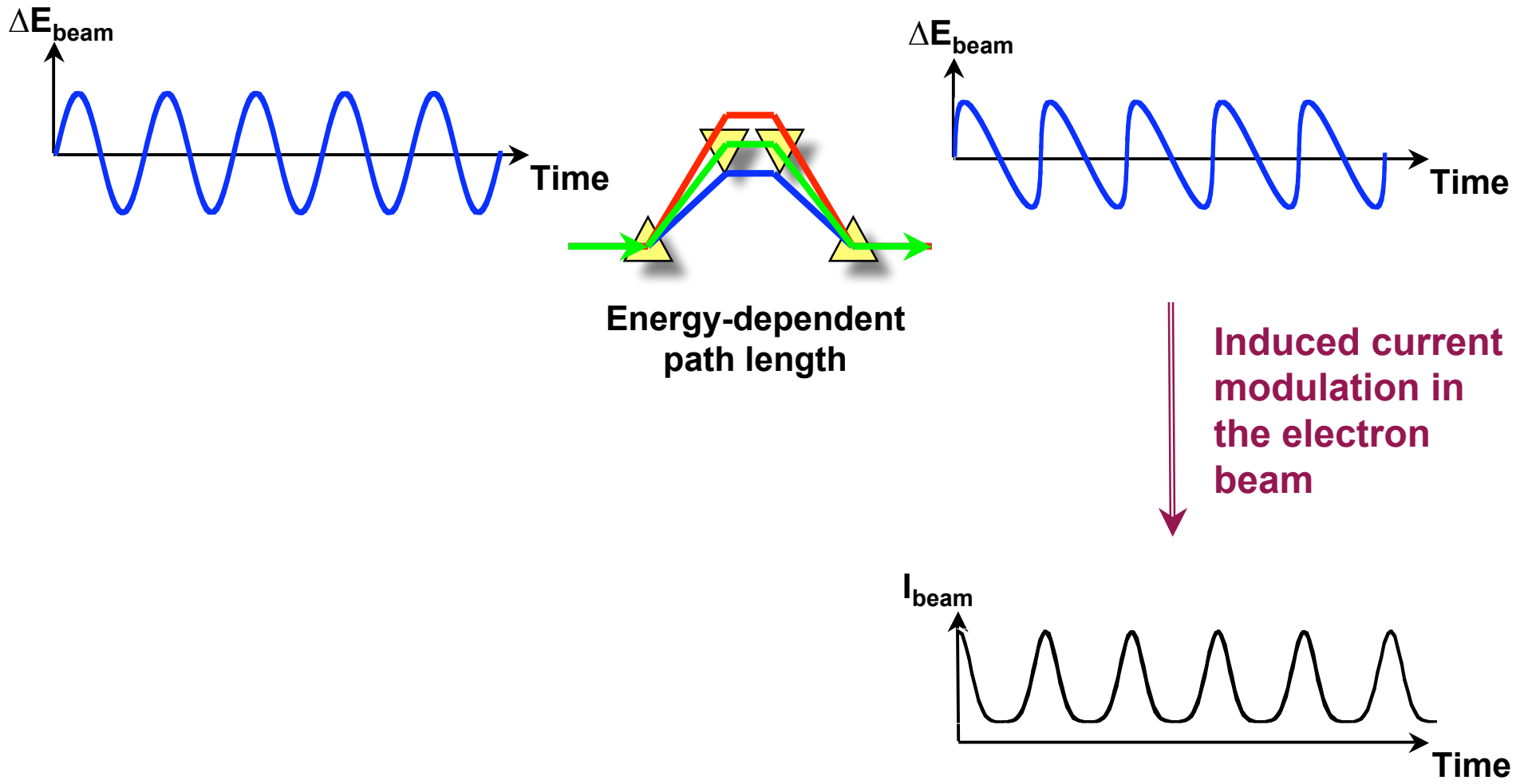


Electron beam is 1.5 GeV, energy spread 100 keV, 250 A current, 0.25 micron emittance; laser seed is 100 kW at 32 nm; undulator period 1 cm



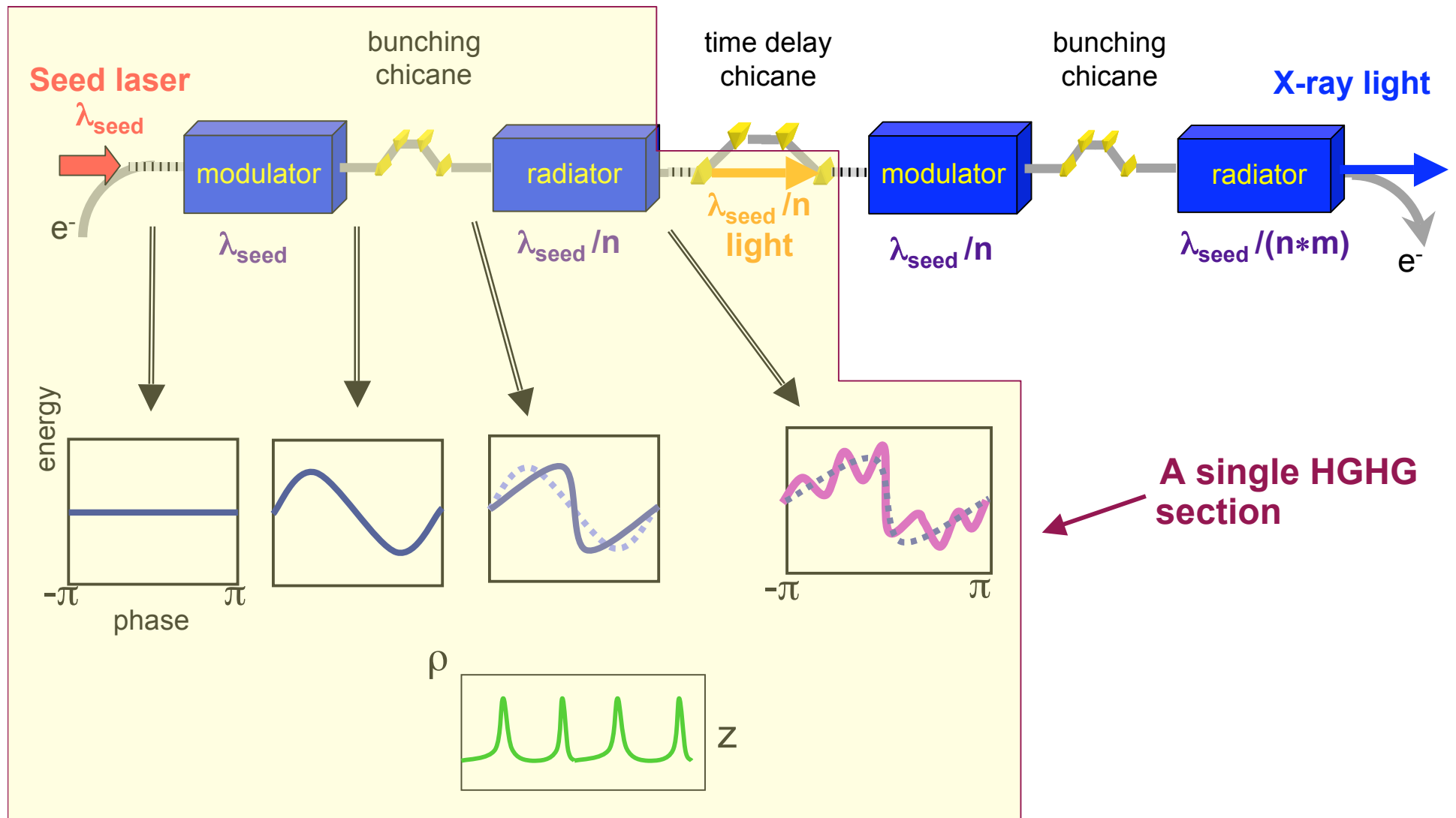
# Bunching of the electron beam

## ENERGY MODULATION FOLLOWED BY DISPERSIVE SECTION



# Harmonic cascade

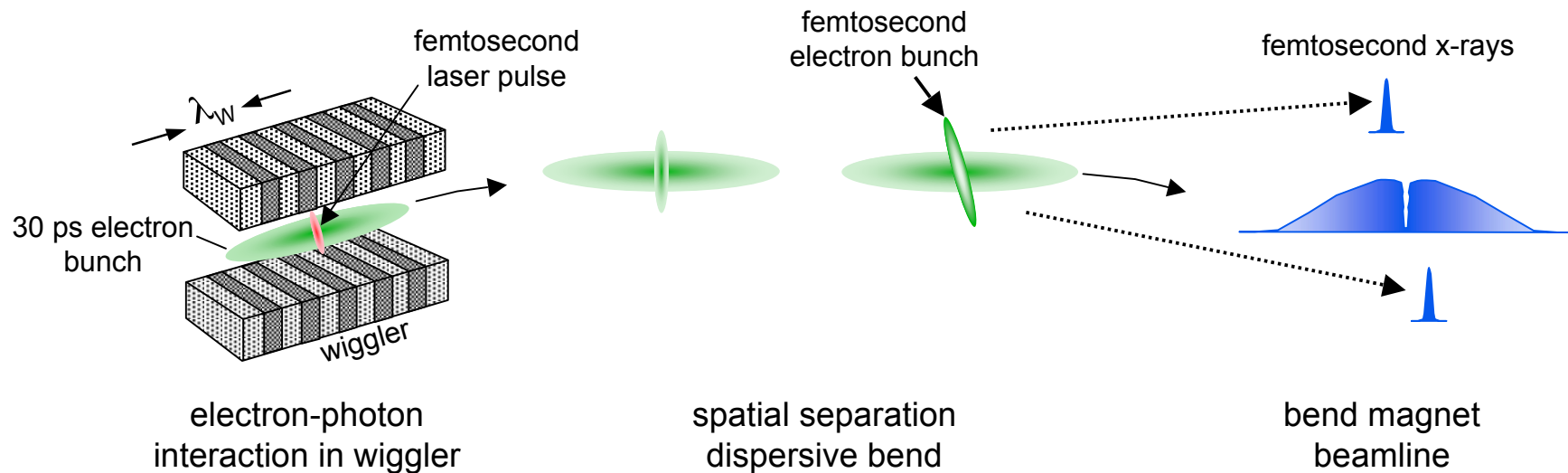
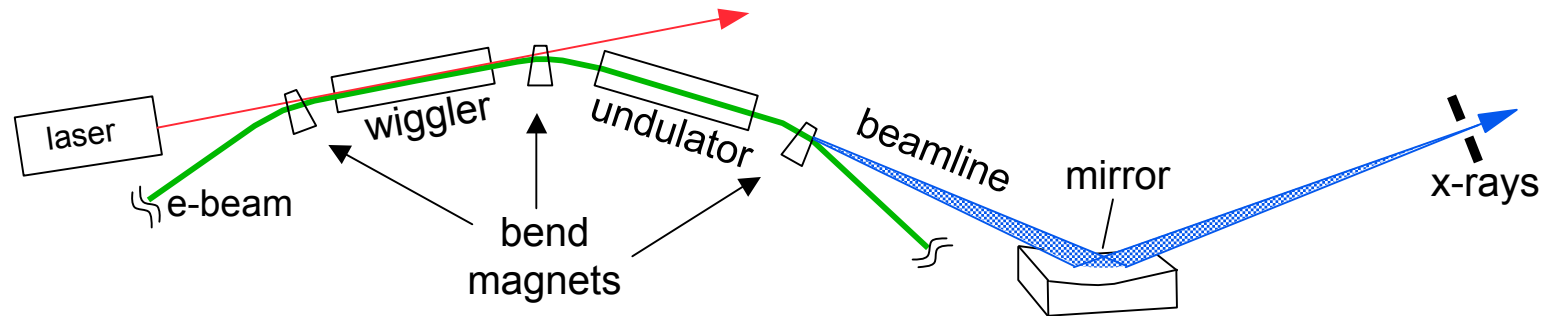
## MULTIPLE STAGES TO REACH SHORTER WAVELENGTHS



Csonka 1980; Kincaid 1980; Bonifacio 1990; L.-H. Yu 1990



## Laser-sliced x-ray pulses from synchrotrons are used as tunable soft and hard x-ray probes

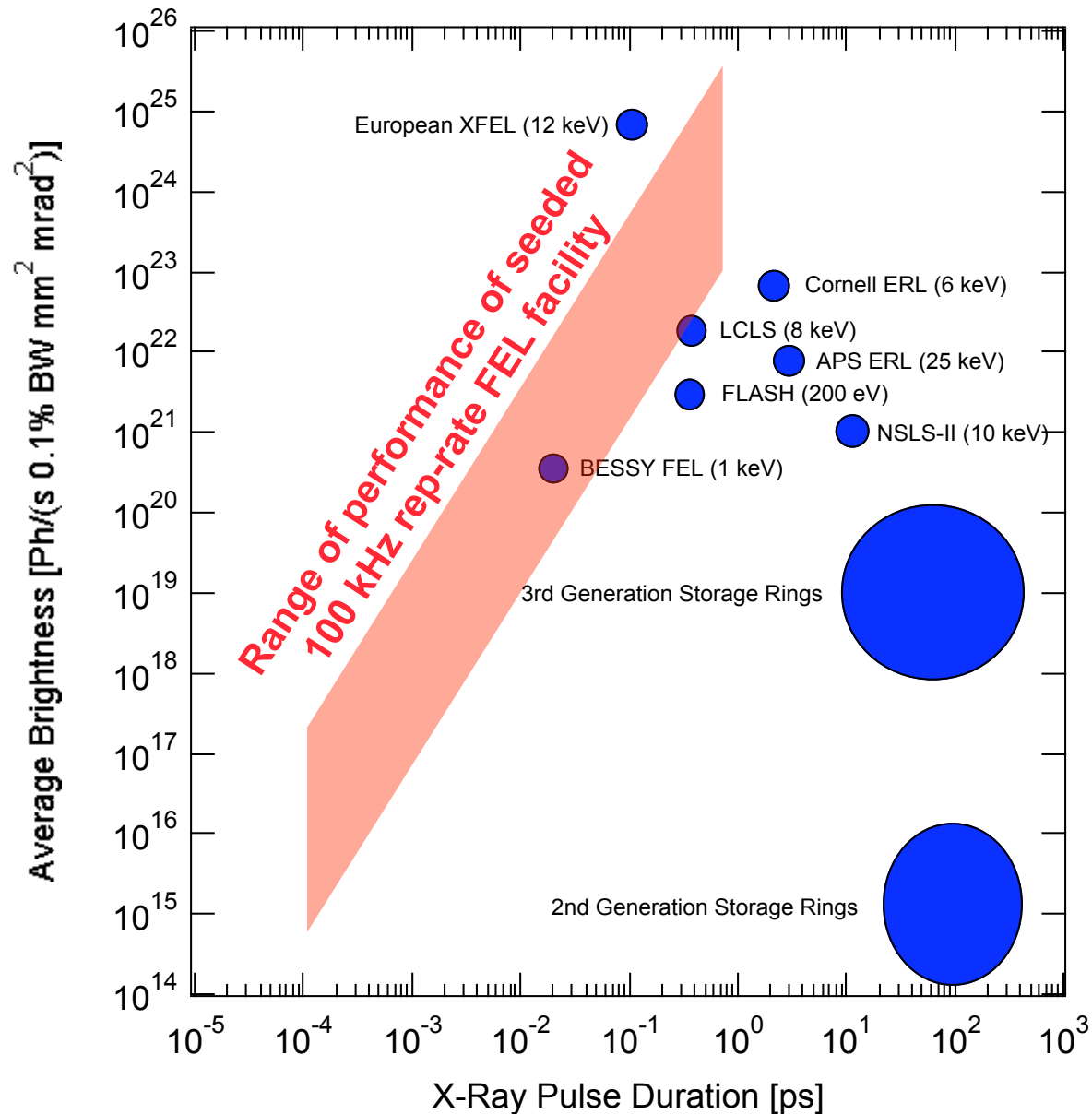


Zholents and Zolotarev, Phys. Rev. Lett., 76, 916, 1996



# New source performance comparison

TIME-DOMAIN RANGING FROM PICOSEC TO SUB-FEMTOSEC





# Summary of integrated photon flux needed in condensed matter physics experiments

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- angle resolved photoemission: volume datasets
  - $1e17$  ph (20 – 100 eV)
- microscopy
  - $1e13$  (280 – 1200 eV)
- spectro microscopy
  - $1e15$  (280 – 1200 eV)
- time resolved microscopy
  - $1e16$  ph (280 – 1200 eV)
- time resolved spectroscopy
  - $1e10$  ph (280 – 1200 eV)

From H. Padmore



## Workshop "High Average Power Lasers and High Harmonics"

December 12, 2007



Discuss future possibilities for high average power lasers that could:

- drive high-peak and high-average power high-order harmonic sources
- be utilized for coherent soft x-ray science
- manipulate electron beams and seed FELs
- enable laser-based accelerators for applications including light sources

# Quasi-phase matching at 300 eV in Ar

PRL **99**, 143901 (2007)

PHYSICAL REVIEW LETTERS

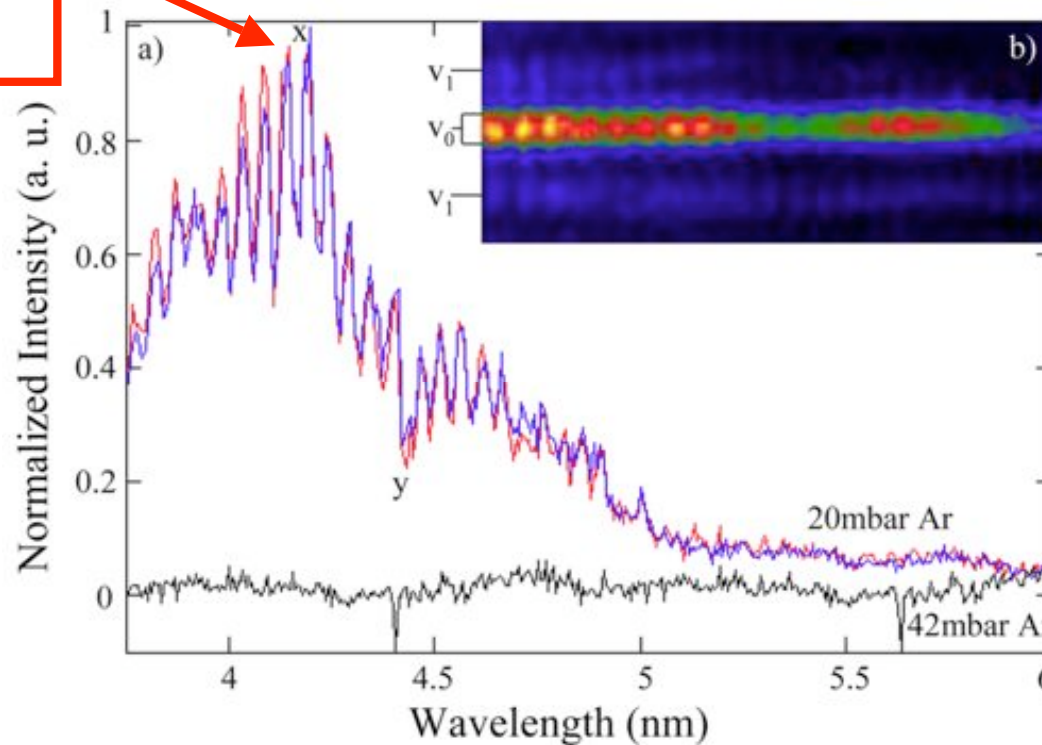
week ending  
5 OCTOBER 2007

## Bright Quasi-Phase-Matched Soft-X-Ray Harmonic Radiation from Argon Ions

M. Zepf,<sup>1,\*</sup> B. Dromey,<sup>1</sup> M. Landreman,<sup>2</sup> P. Foster,<sup>3</sup> and S. M. Hooker<sup>2</sup>

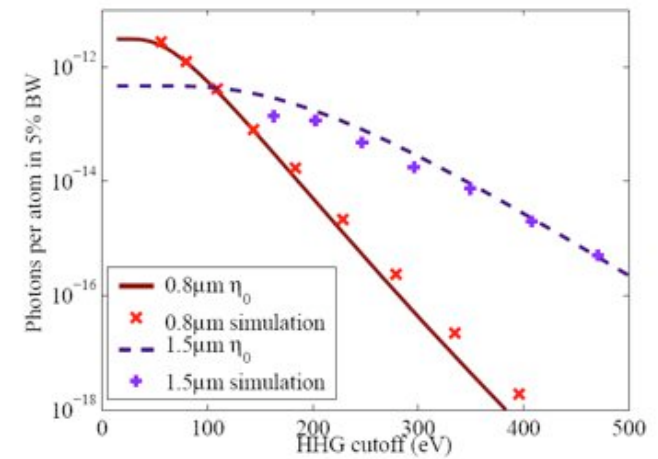
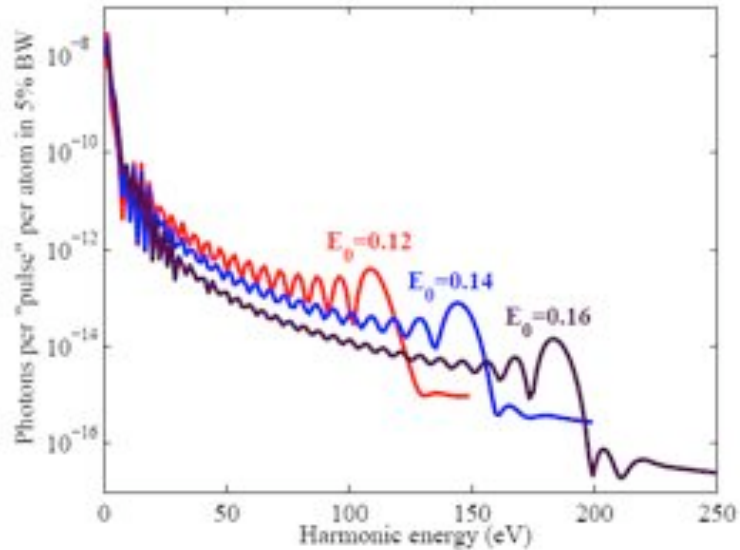
~ E9 photons/pulse/harmonic  
@ 300 eV

~ E12 photons/sec



- 5 mJ / pulse, 800 nm, 40 fsec
- focused to  $\sim 1e15 W/cm^2$  into 1 cm length Ar filled capillary

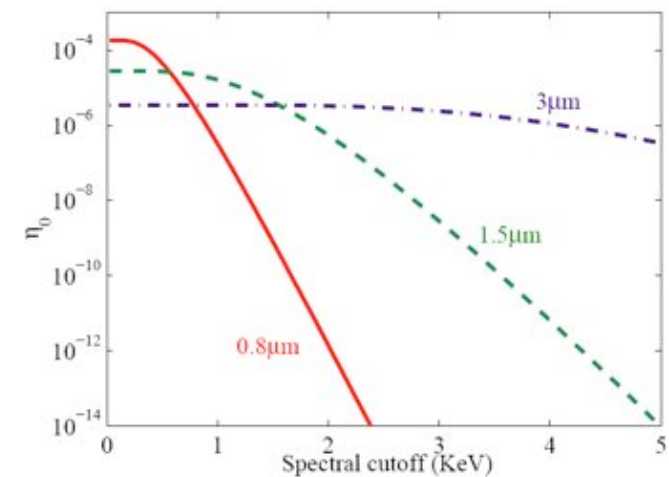
# Optimized HHG at longer drive wavelengths



## Scaling of keV HHG photon yield with drive wavelength

Ariel Gordon and Franz X. Kärtner

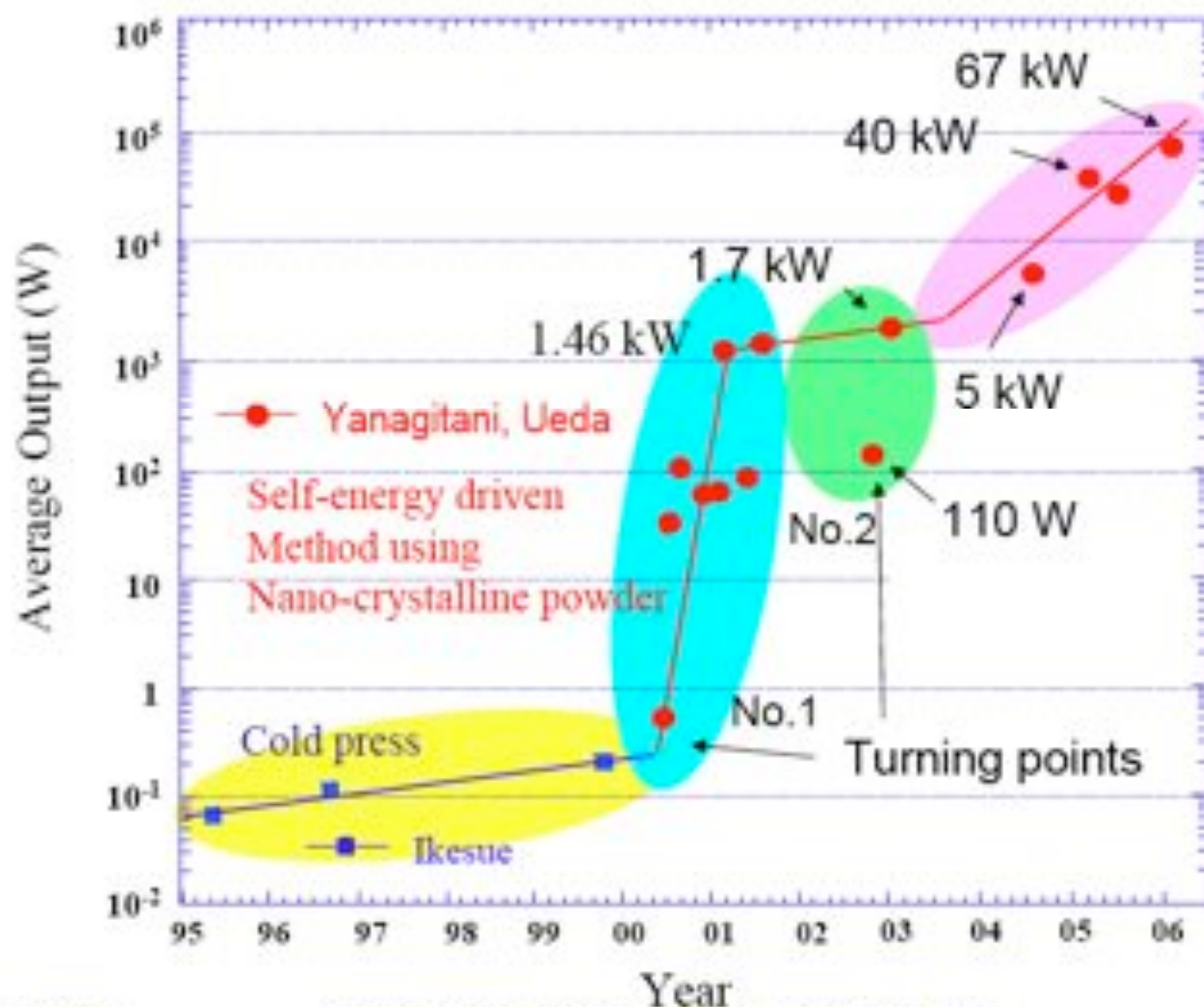
18 April 2005 / Vol. 13, No. 8 / OPTICS EXPRESS 2947





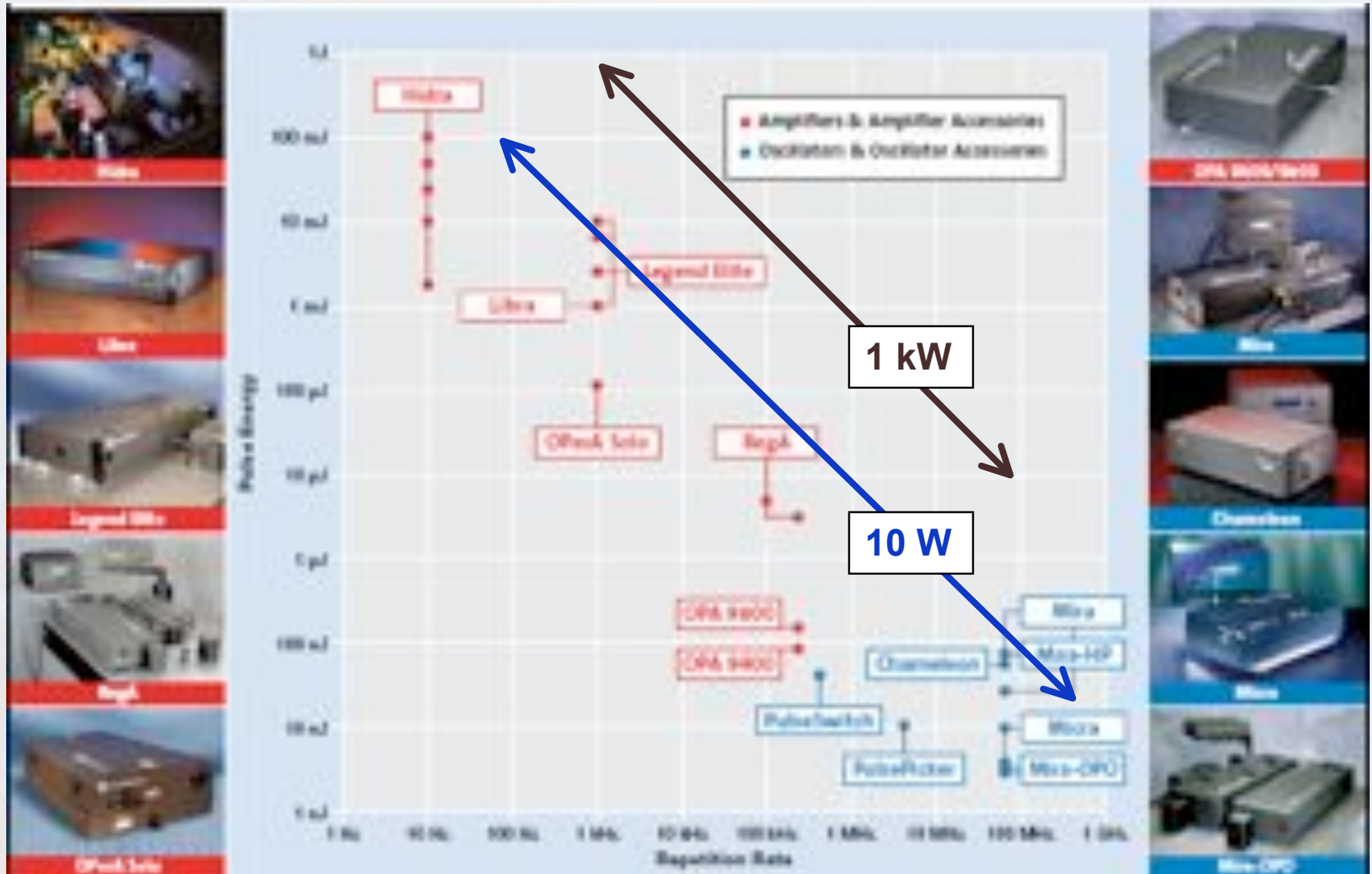
## Rapid Progress of Ceramic Lasers (Ken-ichi Ueda - FMK1 Oct 9 2006)

Byer  
Group



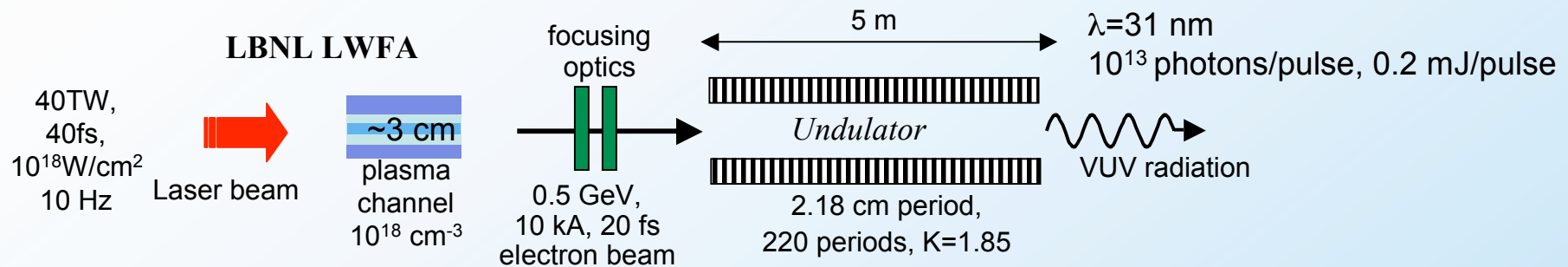


# Largest Selection of Ultrafast Lasers



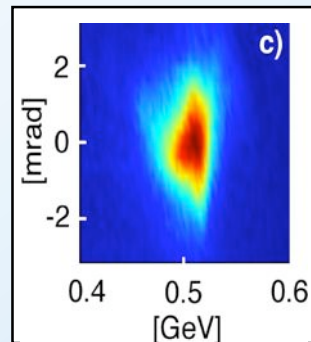


# LWFA-driven coherent VUV source



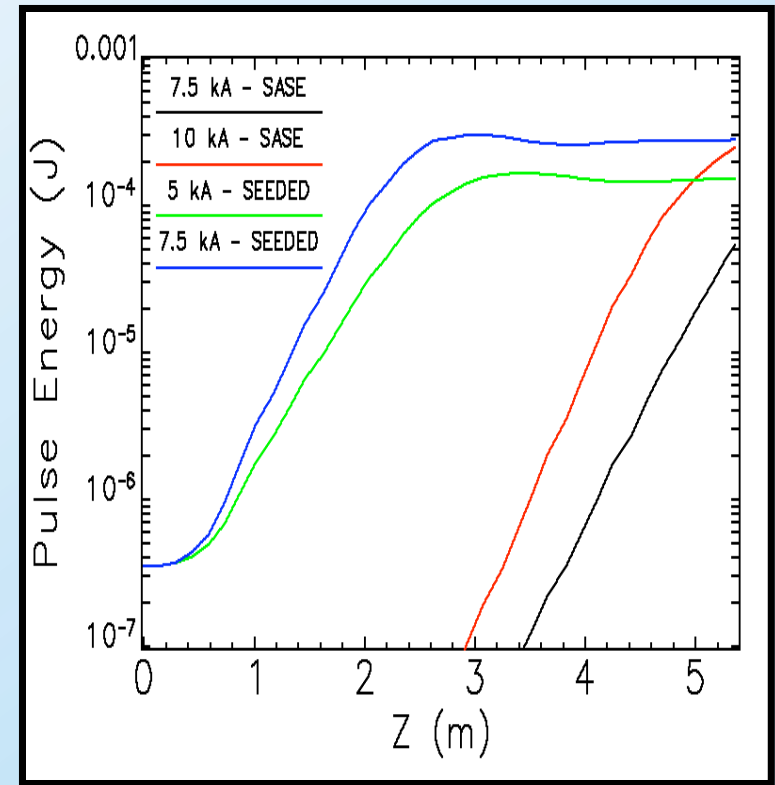
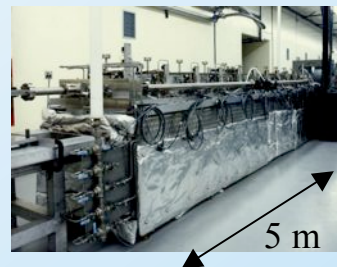
## LWFA Electron Beam:

Beam Energy	0.5 GeV
Peak current	10 kA
Charge	0.2 nC
Bunch duration, FWHM	20 fs
Energy spread (slice)	0.25 %
Norm. Emittance	1 mm-mrad



## Undulator Parameters:

Undulator type	planar
Undulator period	2.18 cm
Number of periods	220
Peak Field	1.02 T
Undulator parameter, K	1.85
Beta function (0.5 GeV)	3.6 m



**Leemans, et al (LBNL) Collaboration  
with MPQ, Germany**

# Conclusions

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- Grand Challenge science requires a range of new x-ray sources
- FLASH, LCLS, the ALS-slicing beamline, APS time-resolved beamlines, etc. are growing new communities of scientists interested in the time domain
- designs exist for high-power x-ray FELs, with flexible parameters and multiple beamlines; R&D is needed
- lasers and high harmonics will have sufficient power for compelling experiments and for seeding coherent, x-ray FELs; R&D is needed
- ... but lasers (even at kW average powers and mW high-harmonics) and crab/slicing sources will not rival average soft and hard x-ray power from FELs, which have mA currents, GeV energies, and watts of coherent x-rays for Grand Challenge experiments
- Crabbing and slicing sources have comparable flux to laser harmonics in the soft x-ray, and greater flux in the hard x-ray
- novel accelerator schemes may eventually become available to drive electron accelerators for light source applications